

Is the ground state of α -(BEDT-TTF) $_2$ MHg(SCN) $_4$ [M =K,Rb,Tl] a charge-density wave or a spin-density wave?

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(February 1, 2008)

The nature of the low-temperature phase of the quasi-two-dimensional conductors α -(BEDT-TTF) $_2$ MHg(SCN) $_4$ [M =K,Rb,Tl] is considered. It is argued that the magnetic field dependence of the phase diagram is more consistent with a charge-density wave, rather than a spin-density wave ground state. The phase diagram of a charge-density wave in a magnetic field is discussed using a Ginzburg-Landau free energy derived from microscopic theory. New experiments are proposed to test the charge-density-wave hypothesis and detect an additional high field phase.

I. INTRODUCTION

Charge transfer salts based on the bis-(ethylenedithia-tetrathiafulvalene) (BEDT-TTF) molecule are novel quasi-two-dimensional conductors.¹⁻³ The widely studied family α -(BEDT-TTF) $_2$ MHg(SCN) $_4$ [M =K,Rb,Tl] have a rich phase diagram depending on temperature, pressure, uniaxial stress, and magnetic field: metallic, superconducting, and density-wave phases are possible.² Band structure calculations show they have co-existing quasi-one-dimensional (open) and quasi-two-dimensional (closed) Fermi surfaces.⁴ At ambient pressure they undergo a transition at a temperature T_{DW} (see Table I) from a metal into a phase that is believed to be a density-wave (DW).⁵ There is currently controversy as to whether this is a spin-density wave (SDW) or a charge-density wave (CDW).⁶⁻⁸ The purpose of this paper is to argue that the field dependence of the phase diagram is more consistent with a CDW than a SDW.

Anomalies in the temperature dependence of the resistivity,⁹ Hall coefficient,¹⁰ specific heat,¹¹ thermal expansion¹² magnetic susceptibility,^{13,14} and muon spin relaxation⁶ suggest there is a phase transition at a temperature T_{DW} from a metal to a different phase. It has been suggested that this is a spin-density wave because (i) the magnetic susceptibility is anisotropic below T_{DW} ^{13,14} and (ii) the muon spin relaxation changes below T_{DW} .⁶ The magnitude of the relaxation implies a magnetic moment of $0.003 \mu_B$.⁶ However, unlike in other SDW systems such as (TMTSF) $_2$ PF $_6$ (which has a magnetic moment of order $0.1 \mu_B$ ¹⁵), no muon spin rotation is observed and nuclear magnetic resonance¹⁶⁻¹⁸ and electron spin resonance¹⁹ measurements show no line broadening or splitting below T_{DW} . The absence of these anomalies could be consistent with the small magnetic moment.

As a magnetic field is applied T_{DW} decreases. At a field H_K (see Table 1), known as the kink field, the low temperature phase is destroyed. The evidence for this is that at H_K qualitative changes occur in angle-dependent magnetoresistance oscillations (AMRO)²⁰ and magneto-oscillations in the resistance,²¹ magnetization and torque.²² At low temperatures the transition at H_K becomes first-order and hysteresis is observed.

II. CDW VS. SDW

If the low-temperature state is a SDW its destruction in high magnetic fields cannot be explained in terms of mean-field theory. The SDW was proposed to result from the nesting of the quasi-one-dimensional Fermi surface. However, in a SDW, unlike a CDW, this nesting is unchanged by the Zeeman splitting of the up and down spin electrons (see Fig. 1). The orbital motion of the electrons in a magnetic field can actually improve the nesting and thus increase the transition temperature.²³ To overcome this problem that the SDW is destroyed at high fields it has been proposed that magnetic breakdown²⁴ or the field enhancing quasi-one-dimensional SDW fluctuations²⁵ are responsible for the destruction of the SDW. However, both these effects involve the orbital motion of the electrons and so imply that when the field is tilted at an angle θ away from the normal to the most-conducting planes that the kink field should vary as $H_K(\theta = 0)/\cos\theta$. This is not observed: $H_K(\theta)$ depends very weakly on θ for $\theta < 50$ degrees.^{8,22,26,27}

III. CDW IN A MAGNETIC FIELD

If the low-temperature phase is a CDW it is not difficult to explain its destruction at high fields. I now briefly consider the essential physics of the theory of a CDW in a magnetic field.²⁸⁻³² For simplicity, only the case of a strictly one-dimensional system is considered and treated at the mean-field level. The only effect of a magnetic field

H is to introduce a Zeeman splitting $2\mu_B H$, where μ_B is the Bohr magneton, between the energies of up and down spin electrons. For simplicity the effects of fluctuations, orbital motion, and the quasi-two-dimensional Fermi surface are neglected. The Zeeman splitting degrades the nesting of the Fermi surface (Fig. 1) and reduces the mean-field transition temperature until it goes to zero (Fig. 2).

There is a simple physical argument to explain the destruction of the CDW by a large field. It is the analogue of an argument used to describe the Pauli or Clogston-Chandrasekhar limit for superconductors.³³ (This is important when orbital effects can be neglected such as in thin films in a magnetic field lying in the plane of the film.³⁴) In a magnetic field the metallic state gains an energy $-\chi H^2$ where χ is the Pauli spin susceptibility, $\chi = \mu_B^2 \rho(E_F)$ and $\rho(E_F)$ is the density of states at the Fermi energy. However, at zero temperature formation of a CDW lowers the total energy of the system by about $-\rho(E_F)\Delta(0)^2$. Hence, at fields larger than $H \simeq \Delta(0)/(\sqrt{2}\mu_B) = 1.2k_B T_{CDW}(0)/\mu_B$ the CDW will be unstable. The results of detailed microscopic calculations are consistent with this rough estimate (compare Fig. 2).

At low enough temperatures and high enough fields it is favourable to form a new CDW phase, denoted CDW_x , with a wavevector shifted away from $2k_F$, the wavevector in zero-field. The shift in wavevector increases with field. This phase is the analogue of the Fulde-Ferrell phase that is predicted to occur in superconductors (with suppressed orbital effects) in a high magnetic field (or strong spin exchange)^{35–43} and the analogue of the incommensurate phase that occurs in spin-Peierls materials in a high field.^{44–47} The mathematics describing these three different systems is almost identical.

IV. GINZBURG-LANDAU THEORY

To clarify the physics leading to the phase diagram shown in Fig. 2 it is helpful to formulate the description of the CDW transition in terms of Ginzburg-Landau theory. The simplest Ginzburg-Landau free energy functional $F[\phi]$ for a one-dimensional system with a complex⁴⁸ order parameter $\phi(x)$, where x is the spatial co-ordinate, is

$$F[\phi] = \int dx \left[a |\phi|^2 + b |\phi|^4 + c \left| \frac{\partial \phi}{\partial x} \right|^2 \right] \quad (1)$$

where the coefficients a , b , and c can be derived from microscopic theory (see the Appendix). In order to have a stable ground state b and c must be positive. The mean-field transition temperature is defined by the temperature at which the second-order coefficient $a(T)$ vanishes. This description is valid at low fields. T_{CDW} decreases quadratically at low fields. However, at $\mu_B H = 1.9101k_B T$ both the coefficients b and c vanish signalling that new physics becomes important. (The fact that both b and c vanish simultaneously is a consequence of weak-coupling theory and presumably does not occur in more general situations).

Generally, b becoming negative denotes the transition becoming first order⁴⁹ and c becoming negative denotes the appearance of a modulated (i.e., spatially non-uniform) phase⁵⁰. To treat these effects the Ginzburg-Landau functional should be expanded to higher orders in the order parameter and its gradients.

$$F[\phi] = \int dx \left[a |\phi|^2 + b |\phi|^4 + e |\phi|^6 + c \left| \frac{\partial \phi}{\partial x} \right|^2 + d \left| \frac{\partial^2 \phi}{\partial x^2} \right|^2 \right] \quad (2)$$

The coefficients d and e must be both positive in order for the ground state to be stable.

We now consider only a single Fourier component, i.e., assume $\phi(x) = \phi_x \exp(iqx)$ (this is also known as the helical state) and minimize the free energy with respect to both q and ϕ_0 . If q is small, then $a(q) \equiv a + cq^2 + dq^4 + \dots$. The optimum wavevector q_x is then determined by

$$a'(q_x) = 0 \quad (3)$$

and the amplitude of the order parameter is determined by the minimum of

$$F[\phi_q] = a(q_x) |\phi_x|^2 + b(q_x) |\phi_x|^4 + e(q_x) |\phi_x|^6 \quad (4)$$

The transition between the uniform (metallic) and the CDW_0 phase is determined by $a(0) = a = 0$. Contrary, to the assumptions of some authors this equation does *not* determine the boundary between the CDW_0 and the CDW_x phases. The exact location of that boundary is determined by finding where the free energy of these two phases is equal. However, $a = 0$ does give its approximate location. Since $b < 0$ for $\mu_B H > 1.9101k_B T$, the transition from the CDW_0 to the CDW_x phase is *first order*, contrary to the claims of some.^{31,32} One also then needs to calculate the regions of metastability of these two phases. The boundary between the uniform (metallic) and CDW_x phase

is determined by $a(q_x) = 0$. I now show that this transition is *second order*, contrary to what has been claimed.³² Microscopic theory implies the identity $b(q) = \frac{\partial^2}{\partial q^2} a(q)$.³⁹ Hence, for small q_x , $\frac{\partial}{\partial q} a(q_x) = 2q_x(c + 2dq_x^2) = 0$ and thus $b(q_x) = 10dq_x^2 > 0$.

V. COMPARISON WITH EXPERIMENT

Although there are numerous quasi-one-dimensional materials with a CDW ground state most have transition temperatures of the order of 100 K and so fields of the order of several hundred tesla would be required to destroy the CDW. However, $\text{Per}_2[\text{Au}(\text{mnt})_2]$ has a transition temperature of 12 K and was recently studied at fields up to 18 tesla.⁵¹ It was found that the transition temperature decreased quadratically with field, but at only half the rate predicted by the mean-field theory discussed here.

The phase boundary of the CDW_0 phase shown in Fig. 2 is quantitatively consistent with the phase boundary observed for the DW phase of $\alpha\text{-(BEDT-TTF)}_2\text{MHg(SCN)}_4$ [$M=\text{K, Rb, Tl}$]. The transition becomes first order for $T < 0.4T_{DW}(0)$. Table I shows that the dimensionless ratio $\mu_B H_K / k_B T_{DW}(0)$ is the same for all three salts. It is about twice the value of 0.9 predicted by Fig. 2. However, it is quite likely that fluctuations reduce $T_{DW}(0)$ below its mean-field value,⁵² thus increasing the ratio $\mu_B H_K / k_B T_{DW}(0)$. If instead of $T_{DW}(0)$ the value of the zero-temperature gap $\Delta(0) = (5 \pm 1)$ meV, estimated from magnetoresistance measurements⁵³ is used then $\mu_B H_K / \Delta(0) = 0.3$, compared to the theoretical value of 0.5.

VI. THE MODULATED PHASE

If the phase diagram of Fig. 2 describes $\alpha\text{-(BEDT-TTF)}_2\text{MHg(SCN)}_4$ [$M=\text{K, Rb, Tl}$] then one should be able to observe the modulated phase. At low temperatures significant hysteresis is observed near the kink field. This is consistent with the transition between the uniform CDW and modulated CDW phases being first order. (Although it could also be glassy behaviour because the CDW wave vector won't line up).

There is some experimental evidence for a phase transition at fields above H_K . First, torque measurements in the K salt show hysteresis near 27.2 T, in addition to the hysteresis at the kink field, of about 23 K.²² Secondly, the character of Shubnikov - de Haas (SdH) oscillations changes significantly on lowering the temperature at fields well above H_K .⁵⁴ Conventional SdH oscillations increase in amplitude with decreasing temperature. In contrast, in the K salt, for fields in the range 36 to 43 tesla, the amplitude of the oscillation at the fundamental frequency increases with decreasing temperature and then decreases until it has a minimum near 1 K and then increases as the temperature is lowered further. These results were interpreted in terms of competition between transport involving the bulk and the surface of the sample. An alternative explanation is that the minimum at 1 K is associated with a phase transition. On the other hand, it should be pointed out this anomaly was not seen in de Haas - van Alphen experiments. Third, recent temperature sweeps of resistance and torque at fixed fields in the range 25 to 28 tesla (and thus above H_K) show features near 4 K.⁵⁵ These features have been interpreted as being due to a transition into a new low-temperature high-field phase. A second set of torque and resistance measurements⁵⁶ has been used to justify an even more complicated phase diagram.

It should be pointed out that the modulated phase may have a much smaller effect on transport properties than the CDW_0 phase. At low temperatures the order parameter decreases by a factor of about three on crossing from the CDW_0 phase to the CDW_x phase (compare Fig. 7. in Ref. 39). If there is a reconstructed quasi-two-dimensional Fermi surface due to the CDW (this is still controversial) then the associated energy gap is proportional to the CDW order parameter,⁵³ and so will also be reduced by three. Since the magnetic breakdown field H_0 for the reconstructed Fermi surface is proportional to the square of the energy gap⁵⁷, H_0 will be reduced by an order of magnitude. In the CDW_0 phase $H_0 \sim 60$ T and so in the CDW_x phase $H > 5H_0$ and the magnetic breakdown will be so complete that the reconstructed Fermi surface will have little effect on transport (compare Fig. 3 in Ref. 53). However, if the magnetoresistance in the density-wave phase is not due a reconstructed quasi-two-dimensional Fermi surface, but due to an alternative mechanism, such as proposed by Yoshioka⁵⁸ then the CDW_x phase might still have an observable effect on the magneto-transport.

Perhaps the transition into the modulated phase from the metallic phase could be detected by measurements of thermodynamic quantities such as specific heat and thermal expansion. The anomaly at low fields is very small¹¹ and will be even smaller for the modulated phase. Yet hopefully, it can still be detected on high-quality crystals. For the spin-Peierls compound CuGeO_3 the transition into the modulated phase was recently detected by thermodynamic measurements.⁵⁹

VII. FUTURE DIRECTIONS

In conclusion, it is argued that the magnetic field dependence of the phase diagram of α -(BEDT-TTF)₂MHg(SCN)₄[M=K,Rb,Tl] is more consistent with a charge-density wave, rather than a spin-density wave ground state. Based on this work I suggest some future directions for both theoretical and experimental work. On the theory side four questions need to be addressed.

(1) What is extent of the metastable regions of the CDW and CDW_x phases? The calculations should be compared to the experimental results which show that on down sweeps of the field hysteresis is observed down to fields a small as half of the kink field.

(2) Is the spatial dependence of the lowest energy modulated phase of the form $\phi_0 \exp(iq_x x)$, assumed here, or could it be $\phi_0 \cos(q_x x)$, or the elliptic function $\phi_0 \text{sn}(q_x x)$? These three possibilities are also known as the Fulde-Ferrell³⁵, Larkin-Ovchinnikov,³⁶ and soliton states,^{46,43} respectively and have been considered for the analogous problem for superconductors and spin-Peierls systems. This might change the order of the transition from the CDW to the CDW_x phase.⁴³

(3) If the ground state is a CDW how does one explain the anisotropic susceptibility and muon spin relaxation that have been interpreted as evidence for a SDW? It has been suggested^{7,27} that as in the purple bronze γ -Mo₄O₁₁,⁶⁰ the anisotropy arises from Landau paramagnetism (an orbital effect). The muon spin relaxation data suggesting a very small magnetic moment might be explained by a coexisting CDW and SDW. A possible mechanism for such a ground state has been considered by Overhauser.⁶¹ It is interesting that (TMTSF)₂PF₆ was thought to be a SDW but recent x-ray scattering measurements suggest a coexisting CDW and SDW.⁶²

(4) Could the ground state of α -(BEDT-TTF)₂MHg(SCN)₄[M=K,Rb,Tl] be a SDW and the high field transition be a spin flop transition?⁶³ Spin flop transitions occur in anisotropic antiferromagnets when the Zeeman energy becomes of the order of the anisotropy energy. This causes a change in the relative orientation of the spins. Typical phase diagrams are qualitatively similar to that shown in Fig. 2.⁵⁰ For this explanation to be plausible a theoretical model must be produced which explains how the reorientation of the spins will change the electronic structure so the AMRO and magneto-oscillations change above the kink field.⁶⁴

I propose several experiments which could help resolve some of the issues raised in this paper.

(1) High resolution x-ray scattering measurements should be done in order to directly observe the CDW in α -(BEDT-TTF)₂MHg(SCN)₄[M=K,Rb,Tl].

(2) In the presence of a CDW one can observe new infrared active phonon modes, sometimes called “phase phonons.”⁶⁵ The appropriate infrared measurements should be made.

(3) Specific heat and thermal expansion measurements should be done in high fields in order to see whether there is a new phase above the kink field.

(4) A search should also be made for the modulated phase in Per₂[Au(mnt)₂]. This will require pulsed magnetic fields of the order of 40 to 60 tesla.

ACKNOWLEDGMENTS

This research was supported by the Australian Research Council and the USA National High Magnetic Field Laboratory which is supported by NSF Cooperative Agreement No. DMR-9016241 and the state of Florida. This work was stimulated by discussions with J.S. Brooks who showed me unpublished results of N. Toyota. Discussions with A. R. Bishop, P. M. Chaikin, J. Eldridge, S. Hill, D. Huse, P. Sandhu, T. Sasaki are gratefully acknowledged. The figures were produced by D. Scarratt.

APPENDIX: GINZBURG-LANDAU COEFFICIENTS

The coefficients in the Ginzburg-Landau free energy can be calculated from microscopic theory using standard Greens function techniques.^{38,39,52} For completeness I give some of the expressions here ($k_B = 1$).

$$a(q) = \frac{1}{\pi v_F} \left[\ln \left(\frac{T}{T_{MF}} \right) + \Psi \left(\frac{1}{2} \right) - \frac{1}{2} \sum_{\alpha=\pm 1} \Psi \left(\frac{1}{2} + \frac{\alpha v_F q + 2\mu_B H}{4i\pi T} \right) \right] \quad (\text{A1})$$

where $\Psi(z)$ is the digamma function and v_F is the Fermi velocity.

$$b(0) = \frac{-1}{4\pi v_F} \frac{1}{(2\pi T)^2} \Psi'' \left(\frac{1}{2} + \frac{\mu_B H}{2i\pi T} \right) \quad (\text{A2})$$

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order parameter will be real. This will produce some change in the analysis that follows (e.g. the modulated phase cannot involve the helical state) but the overall physical picture including the phase diagram will be similar.

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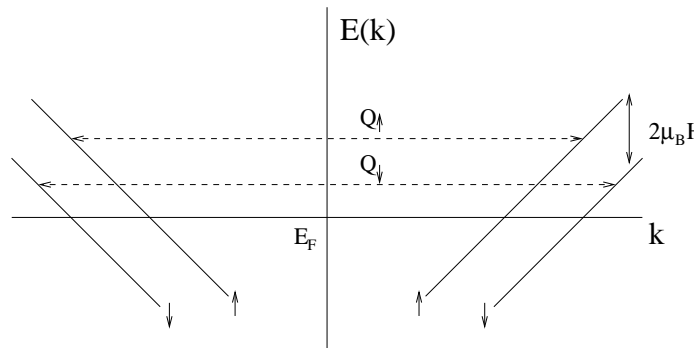


FIG. 1. Dispersion relations of the one-dimensional energy bands in a magnetic field. The Zeeman splitting is $2\mu_B H$. Charge-density-wave correlations couple bands of the same spin. Consequently, simultaneous nesting of the up-spin and down-spin bands is not possible (i.e., the nesting vectors shown, Q_\uparrow and Q_\downarrow , are unequal). In contrast, a spin-density wave involves coupling of bands with opposite spin and the nesting is not affected by a magnetic field.

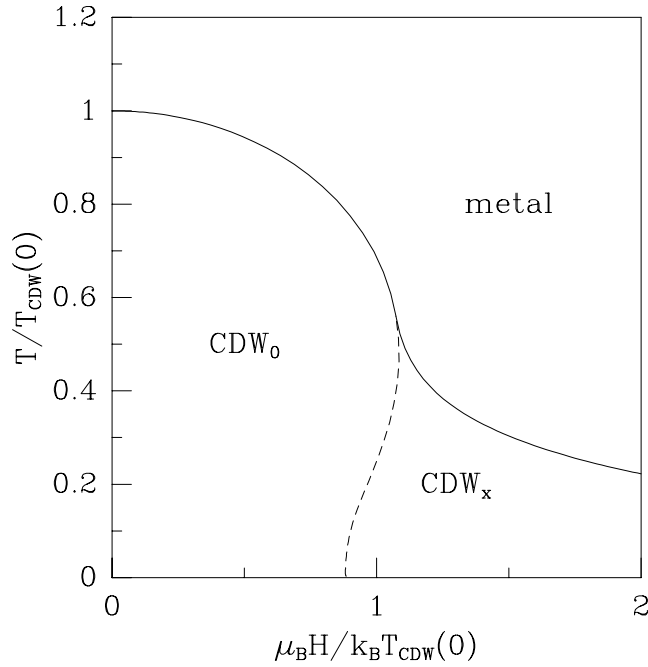


FIG. 2. Phase diagram of a charge-density wave in a magnetic field. The three phases are the metallic phase, charge-density wave (CDW_0) and the modulated phase (CDW_x). The solid line denotes a second-order phase transition. The dashed line denotes the approximate position of a first-order phase transition. In the CDW_0 phase the wavevector $Q = 2k_F$ and is determined by the Fermi surface nesting. In the CDW_x phase the CDW wavevector Q shifts with increasing field. It is an analogue of the Fulde-Ferrell state in superconductors and the incommensurate spin-Peierls state. The temperature T is normalized to the mean-field transition temperature in zero-field, $T_{DW}(0)$.

TABLE I. Values for the zero-field transition temperature $T_{DW}(0)$ and the kink field H_K for the different α -(BEDT-TTF) $_2$ MHg(SCN) $_4$ [M =K,Rb,Tl] salts. The dimensionless ratio $\mu_B H_K / k_B T_{DW}(0)$ can be compared to the value of 0.9 corresponding to the low-temperature boundary of the CDW_0 phase in the phase diagram shown in Fig. 2. The actual ratio is expected to be smaller than 0.9 because the actual transition temperature is reduced below the mean-field value by fluctuations^{52,53}.

M	$T_{DW}(0)$ (K)	H_K (T)	$\mu_B H_K / k_B T_{DW}(0)$
K	8	23	1.9
Tl	9	27	2.0
Rb	12	32	1.8